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Microwave regeneration of monoethanolamine aqueous solutions used for CO_2 capture

Francis Bougie, Xianfeng Fan*

Institute for Materials and Processes, School of Engineering, University of Edinburgh, Mayfield Road, Edinburgh, EH9 3JL, United Kingdom

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ABSTRACT

Aqueous amine absorbents are widely used in post-combustion CO_2 capture processes. To strip the CO_2, the solutions are usually heated around 110–140 °C using a steam reboiler located at the bottom of the regeneration column. In this work, microwave irradiation was studied as a new alternative to heat and regenerate CO_2-rich monoethanolamine (MEA) aqueous solutions. Various parameters like the amine concentration, the regeneration temperature and the initial microwave power were modified to assess regeneration efficiencies by microwave heating and to find optimal experimental conditions in term of recovered CO_2 amount and energy consumption.

The heating rate of solutions by microwave was found to be mostly influenced by their heat capacity, viscosity and CO_2 loading. The optimal heating rate was found for the 50 wt% MEA solution and regeneration performance parameters, based on the CO_2 stripped quantity and microwave energy absorbed by the solution, were also found to be optimal at this concentration. Using a 50 wt% MEA solution instead of the well-known 30 wt% gave a 13% increase in the CO_2 cyclic capacity while the regeneration energy consumption decreased by 15% under the optimal conditions tested in this work. These results indicate that the 50 wt% MEA solution seems to offer great potential in a cyclic CO_2 absorption-microwave regeneration process.

1. Introduction

There has been a great concern in last decades about the global climate changes and the number of scientific researches focusing on reducing the anthropogenic emission of carbon dioxide (CO_2) by post-combustion capture processes has burst. Being already a mature technology and implemented in large scale demonstrations, the CO_2 chemical absorption with aqueous alkanolamine solutions is today's best available technology (Bernardo et al., 2009) and the 30 wt% monoethanolamine (MEA) aqueous solution is widely considered as the benchmark solution for this process (Liang et al., 2015). According to Rochelle (Rochelle, 2009), amine scrubbing will probably remain the dominating technology for CO_2 capture up to 2030. This process consists of the circulation of the absorbent solution between a CO_2 absorption column and a desorption one where the solution is heated to strip the CO_2 (Hüser et al., 2017). The regenerated solvent is then sent back for absorption while the CO_2 is compressed and transported for further use or storage.

It has been reported that the main drawbacks of this amine-based CO_2 capture process arise from the large column sizes (Lassagne et al., 2016) and the high energy consumption in the desorption step which is usually carried out around 110–140 °C and accounts for around 70% of the overall operating cost (Bougie and Iliuta, 2010; Tobiesen and Svendsen, 2006). So far, potential solutions to these problems were mainly focussed on the development of: *(i)* smaller and more efficient gas-liquid contactors such as rotating bed or membrane contactors (Bougie et al., 2014; Wang et al., 2015); or *(ii)* new energy-friendly absorbents such as thermomorphic biphasic solvents or sterically hindered alkanolamines (Bougie and Iliuta, 2012; Zhang et al., 2013). Recently, some researchers also investigated the use of acid catalysts (like $SO_4^{2-}/ZrO_2/\gamma\text{-}Al_2O_3$, H-ZSM-5 or SAPO-34) in the desorption process to facilitate the release of CO_2 and to reduce the heat duty (Zhang et al., 2018a, b; Zhang et al., 2017). Here, one new possibility is investigated with the use of microwave irradiation as an alternative approach to the conventional thermal heating to recover CO_2 from rich MEA solutions.

Theoretically, microwave heating is based on the ability of molecules with a dipole moment to absorb microwave energy and effectively convert it into heat. Two constants are basically involved: the dielectric constant (ε') which is a measure of the substance's ability to store microwave energy, and the dielectric loss factor (ε'') which is a measure of the substance's capacity to convert the stored electromagnetic energy into heat (Cherbański and Molga, 2009; Lee et al., 2016). These constants are function of the microwave frequency, temperature and

* Corresponding author.
E-mail address: X.Fan@ed.ac.uk (X. Fan).

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solution composition (through variation of the molecular interactions) (El Khaled et al., 2016). As microwave heating occurs via direct molecular interactions with the electromagnetic radiations, the main advantages are an instantaneous and volumetric heating without the heat transfer restrictions and heat losses associated with the conventional conductive or convective heating modes. It was also found that microwave could be used for specific or directed heating of one component in a mixture based on their different dielectric constants (Sumi and Horikoshi, 2015). Benefiting from the above-cited advantages, many microwave heating applications were developed in the last decades for example in material synthesis, food processing or chemical reactions and several review papers were published on these subjects (Cherbański and Molga, 2009; Estel et al., 2017; Jones et al., 2002; Mishra and Sharma, 2016). Recent publications were even found using microwave irradiation to regenerate solid CO_2 adsorbents (Chronopoulos et al., 2014; Webley and Zhang, 2014; Yang et al., 2015). However, only one very recent research paper (McGurk et al., 2017) was found using microwave irradiation to heat a spent aqueous amine solution to desorb CO_2. That study showed that microwave could regenerate the solution more quickly than with conventional heating and at quite low temperatures (70–90 °C), potentially reducing overall process costs.

There is clearly a lack of research about the use of microwave to recover CO_2 from rich amine aqueous solutions so the present work was accomplished to understand in more detail this new process by studying the microwave regeneration of MEA aqueous solutions. Several parameters that could influence the regeneration efficiency as the amine concentration, the microwave initial power and the stripping temperature were investigated and the released quantities of CO_2 were compared to the energy absorbed by the solution to find the optimal regeneration conditions. At our knowledge, such study is lacking from the literature and is primordial to assess the efficiency of microwave heating for the regeneration of rich amine solutions and to optimize this process.

2. Experimental section

2.1. Reagents

In this work, all aqueous amine solutions were prepared using distilled water and MEA (CAS no. 141-43-5). The amine (from Sigma-Aldrich UK) had a minimum purity of 98% and was used without further purification. A Sartorius ED224S balance with a precision of $\pm 1 \times 10^{-4}$ g was used to prepare the solutions and it was calculated that the uncertainty of the reported concentrations was less than 0.1 wt%. Gases used for absorption and desorption experiments (CO_2 and N_2) were of commercial grade with a minimum purity of 99.99% (Linde Group UK).

2.2. Microwave generator setup

As an exhaustive description of the microwave regeneration setup can be found elsewhere (McGurk et al., 2017), only a succinct one will be given here. Microwave regeneration of the CO_2-loaded aqueous MEA solutions were all performed with a setup, shown in Fig. 1, composed of a microwave magnetron operating at 2.45 GHz and controlled by an Alter SM445 power supply which maximum output power is 1.2 kW. A single mode waveguide directed the microwave energy towards respectively: *(i)* a dual-directional coupler (GAE Inc., GA310x, calibrated to take into account the presence of the quartz reactor) which measure the forward and reflected microwave power flow, *(ii)* a resonant cavity where the liquid sample is located, and *(iii)* a sliding short circuit (Sairem) to reflect the microwave wave at normal incidence. Non-absorbed power was directed and absorbed by a water dummy-load.

In the sample cavity, 5 g of solution was contained in a cylindrical quartz reactor which had an outside diameter of 17 mm and a thickness of 1.5 mm. The temperature of the solution was measured at the center

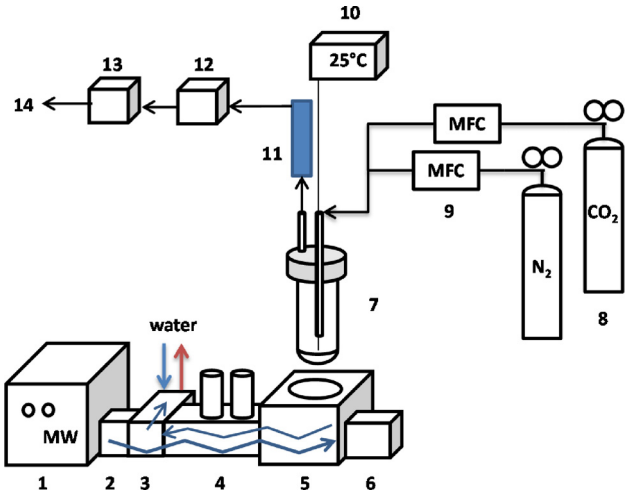

Fig. 1. Experimental setup used for CO_2 absorption and regeneration of the solution by microwave. (1) Magnetron, (2) waveguide, (3) circulator with water dummy load, (4) dual-directional couplers, (5) sample cavity, (6) sliding short circuit, (7) quartz reactor, (8) gas cylinders, (9) mass flow controllers, (10) fiber optic temperature sensor and signal conditioner, (11) water condenser, (12) CO_2 sensor, (13) mass flow meter, (14) gas exit.

of the liquid bulk every second with a fiber optic sensor probe (Opsens OTG-MPK8) suited for use with amine solutions and microwave irradiation. Gases were fed via two calibrated mass flow controllers (Brooks Instruments GF-Series, 0–400 ml/min N_2, 0–100 ml/min CO_2) and bubbled at the bottom of the solution through a small quartz tube. The outlet gas stream of the reactor passed successively through a cold trap, a non-dispersive infrared CO_2 sensor (COZIR-W-100, CO_2 calibration range: 0–70% v/v) and finally a flow meter (Brooks Instruments SLA5860, 0–500 ml/min) before exiting through an exhaust line. All experiments were performed at atmospheric pressure and to avoid unexpected CO_2 absorption by the solution, all gas lines were purged with N_2 before and between experimental runs.

2.3. Absorption procedure

In order to obtain a CO_2-loaded aqueous MEA solution and perform the regeneration, CO_2 absorption was first accomplished by feeding the quartz vial with a binary gas mixture of 20% CO_2 and 80% N_2 which was bubbled through the 5 g MEA solution at a total flow rate of 100 ml/min and at ambient temperature. The outlet CO_2 flow rate was calculated by multiplying the CO_2 sensor and flow meter readings. A blank absorption run was performed with an empty reactor to allow the absorbed quantity of CO_2 to be calculated by integrating the difference between the breakthrough blank and sample CO_2 outlet flow rate curves. The CO_2 loading was calculated as the ratio of the absorbed quantity of CO_2 (mol) to the amount of amine (mol) in solution.

2.4. Regeneration procedure

Once a loaded MEA solution was obtained, the microwave regeneration step could be performed. A N_2 flow was first set at 100 ml/min to purge the gas lines from CO_2 and then acted as a sweeping gas. The microwave source was turned on at an initial power of 100 W (unless stated otherwise) to heat the solution from ambient to the desired regeneration temperature and then the power was manually reduced and controlled to maintain a constant temperature. On the setup, the dual-directional couplers measured the forward and reflected microwave power flows and the difference between them represented the amount of energy absorbed by the solution. Temperature selection in this work (70–90 °C) was based on the results of McGurk (McGurk et al., 2017) who showed that microwave regeneration was effective and

quite fast at these temperatures. Higher temperatures were not selected to keep solvent evaporation rate as low as possible and to avoid significant solvent loss (which could be quite detrimental for a small sample as solution concentration can change over time). Furthermore, an optimized process operating at a low temperature will allow reducing heat losses as well as solvent degradation and corrosion problems during an industrial application. At the end of the regeneration time, the magnetron was turned off and the solution was allowed to cool down. The amount of stripped CO_2 was determined by direct integration of the outlet CO_2 flow rate which was calculated by multiplying the CO_2 sensor and flow meter readings.

3. Results and discussion

3.1. Heating profile

Before performing regeneration analysis of CO_2-loaded MEA solutions, fresh solutions were first submitted to microwave irradiation to gauge their behaviour to this specific heating mode. This preliminary work allowed to select an adequate initial microwave heating power of 100 W which was sufficient to heat the solutions to high temperatures (80 °C) in a relatively short time frame (from around 30–50 seconds depending on the MEA concentration). Several MEA concentrations were tested and their heating profile can be seen in Fig. 2a along with the heating curve of pure water. All experiments were performed at least twice and the reproducibility was excellent, giving the same trend every time. It should be noted that all data included in figures in this work can be found in a dataset available online (Bougie and Fan, 2018). From Fig. 2a, it is first possible to see that the heating rate increases with MEA concentration but up to 50 wt% MEA. The 60 wt% MEA solution has a similar heating profile than the 50 wt% one while the 70 wt % MEA solution take more time to reach the temperature set point and heat up at a rate similar to the 40 wt% MEA solution.

These heating rates can be partly explained by the heat capacity of each solution. Data reported in the literature (Chiu and Li, 1999) show that the heat capacity of MEA solution decreases continuously with an increase in concentration at a given temperature (e.g. 4.19, 3.76, 3.43 and 3.14 J/g.K for water and 30, 50 and 70 wt% MEA respectively at 25 °C). Therefore, as the solutions need less energy, the heating rates should increase continuously in the same way, which is true up to 50 wt % but not for higher concentrations. One possible explanation for the different behaviour at higher concentrations may come from the viscosity of the solution that significantly increases above 50 wt% (e.g. 0.89, 2.48, 5.51, 12.46 mPa.s for water and 30, 50 and 70 wt% MEA respectively at 25 °C) (Amundsen et al., 2009). It was mentioned that molecules in a highly viscous media, or in a media with a high number of intermolecular interactions (which can explain the high viscosity), have a slower response to an oscillating electric field like microwave (Mishra and Sharma, 2016; Salvi et al., 2009). In such media, molecule oscillations will be hindered resulting in a slower rate of volumetric heating. On the other hand, the variation of the heating rate with MEA concentration does not seem to be related to density as this physicochemical property is quite constant (e.g. 0.997, 1.010, 1.021, 1.026 g/cm^3 for water and 30, 50 and 70 wt% MEA respectively at 25 °C) (Tseng and Thompson, 1964).

The heating rates of CO_2-loaded solutions were also determined and results can be seen in Fig. 2b. Several differences can be found in comparison to Fig. 2a; the most obvious one being the longer time

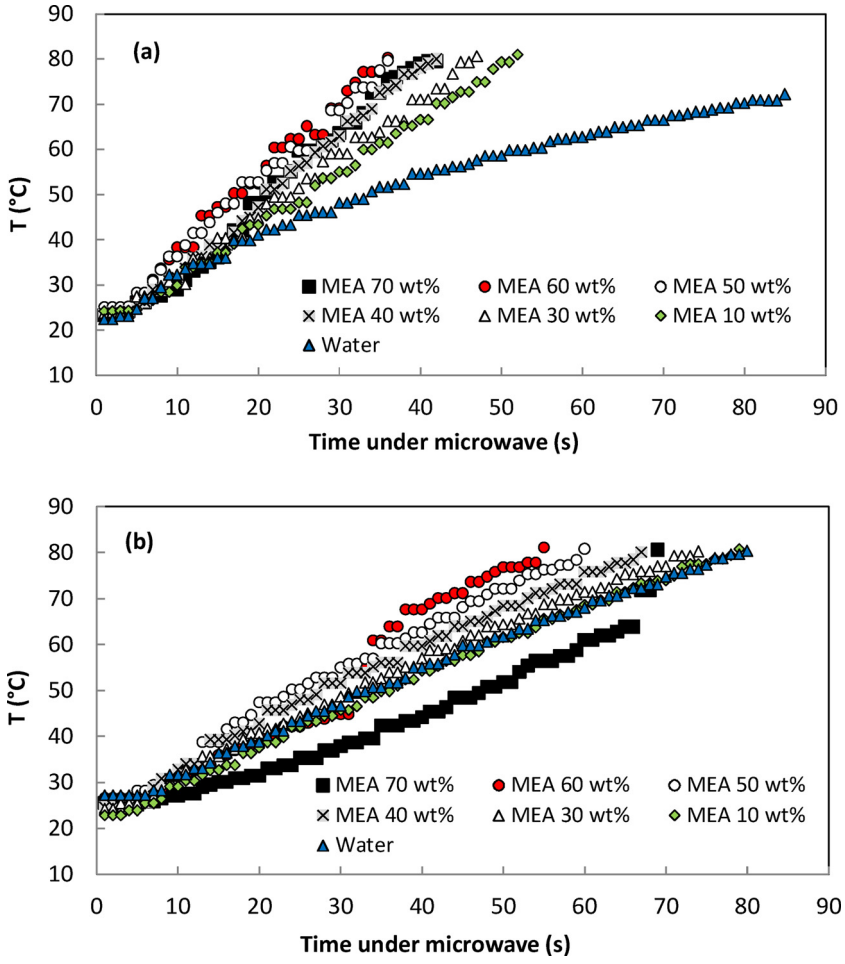

Fig. 2. Heating curves for fresh (a) and CO_2-loaded MEA (b) solutions with a microwave power of 100 W.

needed to reach 80 °C (from 50 to 80 s, depending on the concentration). Again, lower heat capacities for CO_2-loaded solutions in comparison to fresh ones (Weiland et al., 1997) would have alone decreased these time. It means that the viscosity increase after CO_2 absorption (Zhang et al., 2015) has a more significant effect on heating these solutions. For illustration, a 30 wt% MEA solution experiences a decrease of 10% of its heat capacity after reaching a loading of 0.5 mol CO_2/mol amine at 25 °C (Weiland et al., 1997) while its viscosity increases by 46%. The high viscosity of the loaded 60 and 70 wt% solutions was also responsible for the different shape of their heating curve as microbubbles were trapped in the liquid during absorption and released during the heating step, modifying the temperature quickly.

Besides, the slower heating rate of CO_2-rich solutions in comparison to fresh ones can certainly be explained by the endothermic nature of the CO_2 desorption; some microwave energy being diverted to strip the CO_2 instead of heating the solution. One last parameter that may influence the microwave heating rate of these solutions while variating the MEA concentration or the CO_2 loading is the change in their dielectric constants (ε' and ε''). However, as the regeneration of spent aqueous amine solutions is a new field of research, no data were found in the literature to analyse this phenomenon.

Therefore, a maximal heating rate can then be explained to be around 50 wt% MEA based mainly on the opposite trend of the solution heat capacity and viscosity with increasing MEA concentration. At concentrations higher than 50 wt%, the viscosity may become too much detrimental for high microwave efficiency. From a process point of view, a maximal heating rate is interesting as if the solution take less time to reach its temperature set point, a lower residence time in the stripper will be required and the latter can then be smaller.

3.2. Single absorption and regeneration experiments

The heating rate analysis of the previous section demonstrated that MEA concentrations around 50 wt% could be interesting for the CO_2 capture process. Further experiments were then performed to assess the CO_2 absorption and regeneration performance of these solutions.

3.2.1. Absorption and desorption curves

First, the solutions were loaded with CO_2 until a low absorption rate was achieved as it can be seen in Fig. 3. The obtained rich loadings (mol CO_2/mol amine) and corresponding CO_2 absorbed quantity (mol) were indicated directly on that figure for a quick comparison. As expected (Aronu et al., 2011), it can be seen that the loading values decreased with an increase of MEA concentration. This shows that even if there are more amine molecules in a given sample at higher concentrations, these amines are less effective individually to absorb CO_2. Fig. 3 also revealed that a 10 wt% MEA concentration is clearly not enough to maintain a high absorption rate and that a concentration over 30 wt% is required. However, as MEA concentration increased over 50 wt%, the distance between the curves tend to reduce indicating, as in Section 3.1., that a high viscosity increase occurred and that absorption may be limited by mass transfer in the liquid phase. From these loaded solutions, microwave regenerations were performed at three different temperatures: 70, 80 and 90 °C and the CO_2 desorption curves are reported in Fig. 4.

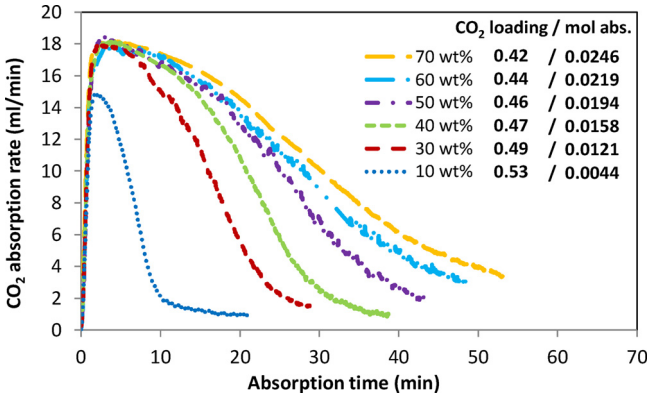

Fig. 3. CO_2 absorption curves for various MEA concentrations from a 20% CO_2 flowrate at 100 ml/min. Achieved loading (mol CO_2/mol amine) and quantity absorbed (mol) are indicated for comparison.

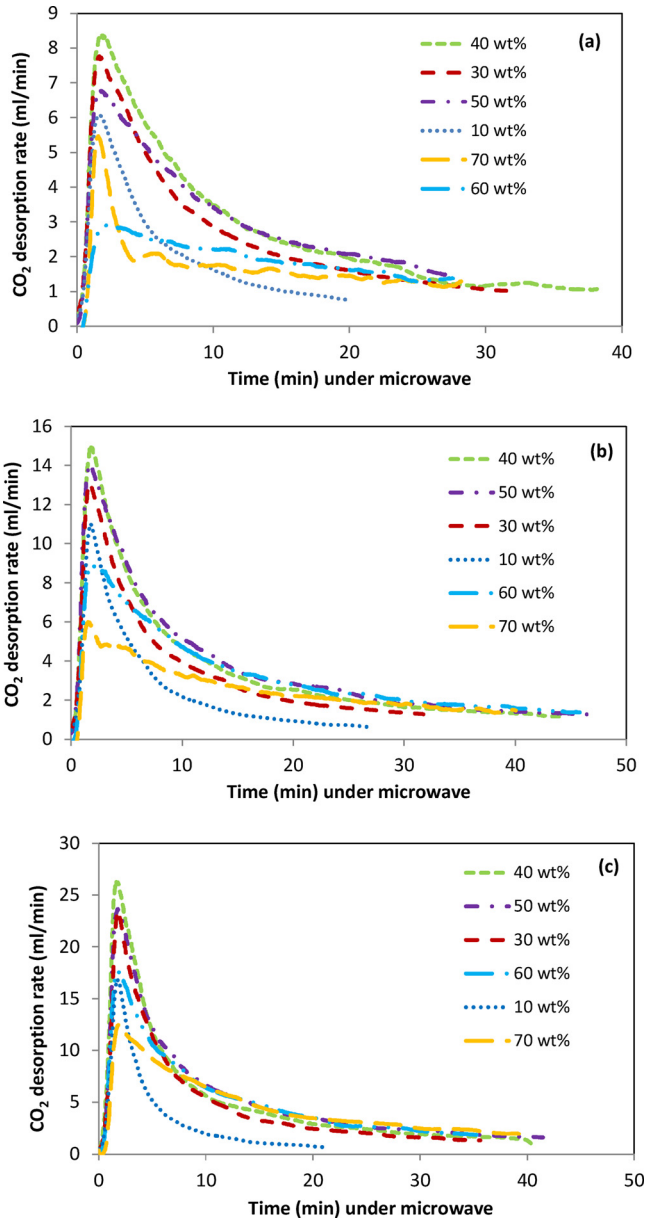

Fig. 4. CO_2 desorption rates obtained performing microwave regeneration at 70 °C (a); 80 °C (b); 90 °C (c).

These regenerations curves were obtained to analyse the efficiency of microwave to release CO_2 at different temperatures and from various MEA concentrations. As the regeneration temperature increase, it is possible to see that the maximal CO_2 desorption rate increase as it could be expected as more energy is supplied to the solution. It is also clearly noticeable that MEA concentrations of 30 wt%, but specially 40 wt% and 50 wt% have the best regeneration performances under all tested temperatures. The integration of these desorption curves can be done to calculate the absolute amount of stripped CO_2 at each condition and for the sake of comparison, the released quantity of CO_2 after 20 min are

Table 1
CO_2 stripped amount after 20 min of microwave regeneration at different temperatures from data of Fig. 4.

MEA	70 °C		80 °C		90 °C	
(wt%)	CO_2 stripped (mol)	% H.A.[a]	CO_2 stripped (mol)	% H.A.	CO_2 stripped (mol)	% H.A.
10	0.0017	53	0.0026	54	0.0029	44
30	0.0031	96	0.0044	90	0.0059	87
40	0.0032	100	0.0047	96	0.0066	98
50	0.0029	92	0.0049	100	0.0068	100
60	0.0017	52	0.0039	81	0.0057	85
70	0.0015	47	0.0027	55	0.0052	76

[a] %H.A.: Percentage of the **H**ighest **A**mount of stripped CO_2 at a given temperature among the different MEA concentrations.

reported in Table 1. As for the desorption rates, the absolute stripped amounts of CO_2 confirmed that MEA concentrations between 30 and 50 wt% perform better at all temperatures. Higher concentrations of 60 and 70 wt% released significantly less CO_2 even though these solutions contained initially more CO_2. As explained earlier, the high viscosity of these high MEA concentrations may explain their poorer regeneration performance. This viscosity effect seems to be validated as when temperature increased, a viscosity reduction is expected resulting in an increase of the % H.A. (as reported in Table 1) for the 60 and 70 wt% MEA concentrations.

3.2.2. Energy consumption analysis

In addition to a high quantity of stripped CO_2, the energy consumption needs to be taken into account in order to determine an optimal MEA concentration to use in an efficient CO_2 capture/regeneration process. As explained in Section 2.4, the microwave setup allowed with the dual-directional couplers to determine the quantity of energy absorbed by the solution during the regeneration steps. It should be mentioned that the desorption energy measured during the regenerations are specific for the setup used in this study and can hardly be compared to values from literature. It is known (Bermúdez et al., 2015) that the microwave desorption energy measured for small samples (as 5 g in this work) overestimate the real energy consumption to strip the CO_2 due, for example, to a high surface/volume ratio of the sample resulting in relatively high heat losses or evaporation rate of the solutions. The heat taken out by the N_2 purging gas and the heat consumed for heating the quartz reactor also interfered. All these energies were not consumed for CO_2 desorption, but are included in the reported data. However, as all experiments are performed under the same conditions, the energy consumptions can be used here to compare solutions of different concentrations to find the optimal one and analyse regeneration parameters.

3.2.2.1. Constant temperature power consumption. Desorption curves of Fig. 4 were all obtained using an initial microwave power of 100 W to quickly heat the solution to the desired regeneration temperature and then the power was manually reduced and controlled to maintain that temperature constant. Values of these constant-temperature powers were recorded and reported in Fig. 5 as some interesting trends can be observed. First, it can be seen that performing the regeneration at higher temperatures required, as expected, higher microwave powers to account for example for higher heat losses from the system, for higher vapour emissions but also for desorbing higher amount of CO_2.

Besides, the analysis of the MEA concentration effect on the power consumption seems to emphasis once again the influence of viscosity on the microwave desorption process. For each tested temperature, the power consumptions were quite similar at low MEA concentrations, as indicated by the horizontal dotted lines in Fig. 5, until a given concentration was reached where the power consumption started to decrease with further concentration (or viscosity) increase. These

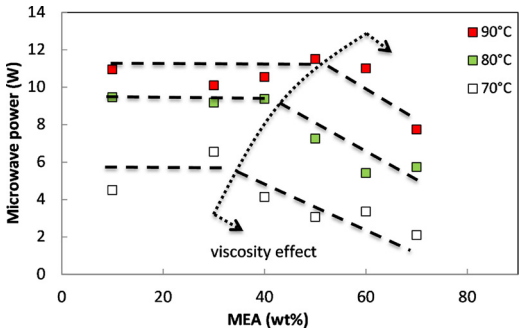

Fig. 5. Constant-temperature microwave powers for various MEA concentrations and temperatures. Dotted lines are included to show trends.

threshold concentrations were found to increase with temperature (around 35 wt% at 70 °C, 40 wt% at 80 °C, and 50 wt% at 90 °C) but more interestingly, they are all linked by the same viscosity value (calculated from literature data (Amundsen et al., 2009)). This indicate that above that viscosity value, the desorption of CO_2 may be hindered by mass transfer limitations resulting in lower desorbed amount and therefore a lower microwave power consumption is necessary.

3.2.2.2. Regeneration performance parameters. In order to perform a regeneration efficiency comparison between solutions, two regeneration performance parameters (*EC and* EC^2) were calculated. Firstly, as shown by Eq. (1), *EC* represent the energy consumption after a given time divided by the amount of CO_2 stripped after the same time (from integration of curves in Fig. 4). Clearly, a low *EC* value is desired as it indicates lower energy consumption per mole of stripped CO_2. Secondly, in the case where several solutions would give similar *EC* values, one more parameter was needed to determine the best solutions. *EC* values were divided by the amount of stripped CO_2 to calculate this new parameter, EC^2, as shown by Eq. (2). A low EC^2 combines two important features: a low *EC* value and a high quantity of stripped CO_2 (needed to have a high cyclic capacity), and can therefore be used as an optimization factor to be minimized in order to find optimal conditions.

$$EC_t = \frac{Energy_t\,(\mathrm{kJ})}{CO_2\ stripped_t\,(\mathrm{mol})} \tag{1}$$

$$EC_t^2 = \frac{EC_t\,(\mathrm{kJ/mol})}{CO_2\ stripped_t\,(\mathrm{mol})} \tag{2}$$

The *EC* results for the three best MEA concentrations at each tested temperature are shown in Fig. 6 while the corresponding EC^2 results are displayed in Fig. 7. The three best concentrations only were included to keep these figures as clear as possible. From Fig. 6, it is possible to see that all the data have a minimum *EC* at the microwave irradiation time around 8 min. The high *EC* values before 8 min can be explained by the

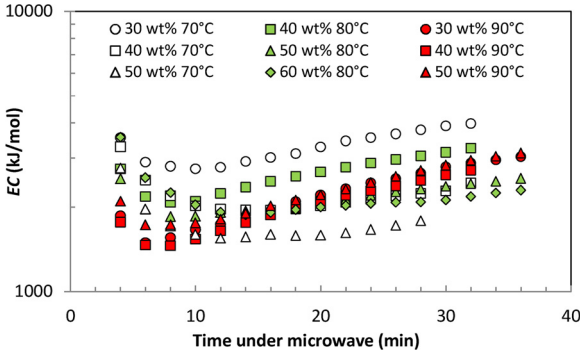

Fig. 6. *EC* as a function of the regeneration time for various MEA concentrations and temperatures.

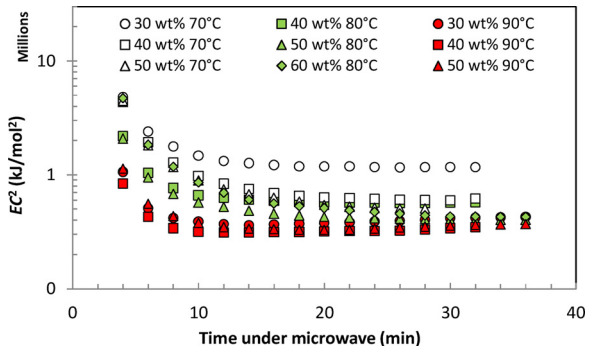

Fig. 7. EC^2 as a function of regeneration time for various MEA concentrations and temperatures.

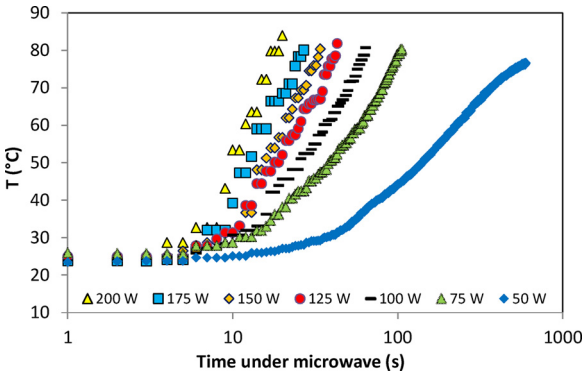

Fig. 8. Heating curves of 50 wt% MEA solutions as a function of initial microwave power during the regeneration steps.

initial sensible heat requirement to heat the solution to the regeneration temperature. Then minimum *EC* values are obtained following the desorption of a high quantity of CO_2. Finally, values start to rise as less CO_2 is released from the solutions (see trends in Fig. 4) while a continuous microwave heating is supplied. Lowest *EC* values were achieved before 10 min of regeneration by solutions at 90 °C while the 50 wt% MEA solution at 70 °C got quite good results for higher regeneration times. The analysis of Fig. 7 confirmed that 90 °C is the best regeneration temperature as EC^2 values are the lowest. The 50 wt% MEA solution at 70 °C did not performed as well as its high EC^2 values emphasis the fact that the amount of CO_2 stripped at 70 °C is significantly lower than the amount stripped at 90 °C after the same irradiation time. The superior regeneration performances, based on *EC* and EC^2, at 90 °C in comparison to the other tested temperatures corroborate the results of McGurk et al. (2017) which were based on solution cyclic loading analysis. This low temperature of 90 °C in comparison to conventional regeneration temperature found in literature (110–140 °C) (Bougie and Iliuta, 2010; Sakwattanapong et al., 2005; Zhang et al., 2008) would be worth considering in a whole process modeling study as solution evaporation rate will be much smaller and amine degradation and corrosion tendency reduced.

3.3. Microwave initial power analysis

So far, results have shown that MEA concentration and regeneration temperature have a significant influence on microwave regeneration efficiency. More experiments were then performed but at different initial microwave powers to see if this factor has also an influence on regeneration performances. For these experiments, 50 wt% MEA solutions were used and submitted to a cyclic process consisting of 20 min absorption steps followed by 10 min regeneration steps at 80 °C until a steady-state CO_2 cyclic capacity was achieved.

Microwave initial powers between 50 and 200 W were tested during the regeneration steps and the effect on the heating rate of loaded solutions at steady-state can be seen in Fig. 8. As expected, a higher power resulted in a higher heating rate of the solutions. A low power of 50 W was not even enough to heat the solution to 80 °C during the allowed 10 min of the regeneration step while a power higher than 200 W was not further considered as a heating time lower than 10 s was achieved and temperature was difficult to control.

Time taken by fresh and loaded solutions to reach 80 °C, their energy consumption during this heating step and the mean absorbed power have been determined and indicated in Table 2. As mentioned earlier, increasing the microwave power resulted in higher heating rates and data in Table 2 also show that, without dissolved CO_2, the energy consumption continuously decreased with the increase in the power up to 150 W and then stayed constant up to 200 W. This can be explained by higher heat losses when the microwave power was low as it took longer time to reach 80 °C. For powers higher than 150 W, the short heating times resulted in similar heat losses and then similar total energy consumption.

Table 2
Energy consumption by 50 wt% MEA solutions to reach 80 °C using different microwave initial powers.

	Fresh solutions			CO_2-loaded solutions		
Nominal microwave power	Energy	Time to 80 °C	Mean absorbed power	Energy	Time to 80 °C	Mean absorbed power
(W)	(J)	(s)	(W)	(J)	(s)	(W)
50	2560	242	11	–	–	–
75	2020	56	36	2697	102	26
100	1534	36	43	1746	62	28
125	1257	26	48	1488	44	34
150	1178	23	52	1288	34	38
175	1203	13	93	1198	23	52
200	1176	10	114	970	18	54

However, for solutions with CO_2, no stabilization in the decreasing energy consumption was found. With the increase of the microwave power, shorter time to reach 80 °C resulted in lower quantity of stripped CO_2 during the heating step so energy requirements keep on decreasing. The energy consumption decreased toward an estimate value of 850 J, which would be the energy requirement to heat the loaded 50 wt% MEA solution to 80 °C without any CO_2 desorption or heat losses as calculated based on heat capacity from literature (Weiland et al., 1997). This can indicate that the primary sink for microwave energy is for bulk solution heating and not for breaking the CO_2-amine bond. Similarly, it is interesting to notice that for powers higher than 175 W, the energy consumption during the heating step for the CO_2-loaded solutions are lower than for the fresh solutions. This is due to the specific heating of the CO_2-loaded solutions (which have lower heat capacity than the fresh ones) without any significant quantity of CO_2 being stripped. These results may point toward the absence of non-thermal microwave effects during the regeneration of the solutions. However, a more detailed study comparing the microwave to conventional regeneration techniques would be needed to confirm this assumption.

Besides, the mean microwave powers absorbed by the solutions during the heating step were also calculated in Table 2 as the energy consumption divided by the heating time. These values give an indication on how much power is absorbed by the solution and how much is reflected (difference between the nominal microwave power and absorbed power). As it could be expected, all mean absorbed power values increased with microwave power and they are all lower than the power emitted by the magnetron. This is due to the fact that the solution in the quartz vial occupies a small cross-section area in the waveguide so some power is not absorbed and goes to the water dummy-load as mentioned in Section 2.2. Also, results tend to confirm, as explained in Section 3.1., that solutions with dissolved CO_2 are less

Table 3
Effect of the initial microwave power on CO_2 cyclic capacity and regeneration performances.

Initial microwave Power (W)	Energy (J)	Cyclic capacity (mol CO_2)	*EC* (kJ/mol)	EC^2 (kJ/mol^2)
50	4930	0.0025	1972	788720
75	6932	0.0035	1981	565882
100	6365	0.0036	1768	491108
125	6311	0.0036	1753	486945
150	6201	0.0035	1772	506204
175	6082	0.0035	1738	496480
200	6020	0.0035	1720	491405

efficient to absorb microwave as they have lower mean absorbed power values than fresh solutions due to their higher viscosity or different dielectric properties.

To complete with the initial microwave power analysis, results from the actual cyclic absorption-microwave regeneration experiments are displayed in Table 3. From these data, a satisfactory initial power can be determined. 50 W is clearly not enough and resulted in a lower CO_2 cyclic capacity and therefore higher *EC* and EC^2 values. On the other side, powers higher than 100 W led to similar regeneration performances. Consequently, the variation of the microwave initial power did not bring significant advantages during these experiments and an initial power of 100 W should be sufficient. With high powers, one can however benefit from a high volumetric heating rate of the rich solutions to slightly reduce energy consumption as CO_2-loaded solutions have lower heat capacities.

3.4. Optimal condition verifications through cyclic experiments

In previous sections, it has been found that MEA concentrations from 30 to 50 wt% offer good CO_2 absorption and desorption rates (see Figs. 3 and 4), that the lowest *EC* and EC^2 values are obtained at 90 °C (see Figs. 6 and 7) and that a high initial microwave power may help to reduce energy consumption due to a fast heating rate of the solutions. For these reasons, experiments including cyclic absorption (23 min) / regeneration (12 min at 90 °C and 200 W initial power) were performed with the above-mentioned solutions. Absorption and regeneration cycles were accomplished until the absorbed amount of CO_2 was the same as the stripped amount, which took usually 3 or 4 cycles to achieve. Regeneration performance indicators (*EC* and EC^2) at steady-state can be seen in Fig. 9 and both values indicate that the 50 wt% MEA solution performed better followed by the 40 wt% and then the 30 wt% MEA solution. Under these optimal conditions, the 50 wt% MEA solution has a 13% higher cyclic capacity while the energy consumption is reduced by 15% in comparison to the well-known 30 wt% concentration. These superior performances can be explained by better CO_2 absorption (more amine molecules per volume of solution) and regeneration (the viscosity of the solution is not too high) rates leading to a higher cyclic capacity and a lower energy requirement (lower heat capacity) for that solution.

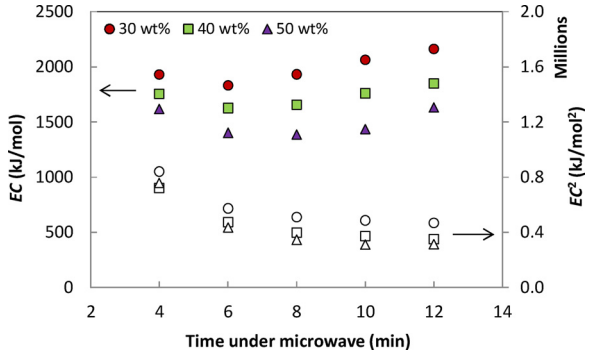

Fig. 9. Regeneration performances of various MEA solutions at 90 °C at steady-state condition during a cyclic absorption/regeneration process.

One can argue that a variation of the CO_2 loading can influence the absorption/desorption rates as well as modify the viscosity, dielectric properties and heat capacity of the tested solutions, therefore changing their cyclic performances. To investigate the effect of the CO_2 loading, other cyclic experiments were performed at 90 °C with the 30 and 50 wt % MEA solutions but using different absorption and regeneration times. A summary of the results obtained at steady-state can be found in Table 4. For the 30 wt% and 50 wt% MEA solutions, rich loadings between 0.18–0.49 and 0.28–0.44 were obtained respectively, showing that a wide range of loading was covered by these experiments. Results indicate that the cyclic capacity decreased when the rich loading was reduced, and that the 50 wt% MEA solution always had a slightly higher cyclic capacity than the 30 wt% solution under the same experimental conditions. These higher cyclic capacities for the 50 wt% MEA solution combined with lower energy consumptions led to better regeneration performance indicators *EC* and EC^2 under all tested conditions.

4. Conclusions

Even if numerous studies were published over the last decades in the literature about the CO_2 capture process by aqueous amine solutions, only one paper was found so far concerning the use of microwave to regenerate the spent solutions indicating that there is still much to learn about this new regeneration technique. In this work, we showed that several parameters impacted significantly the cyclic absorption-microwave regeneration process.

The heating rate of MEA solutions by microwave was found to be influenced by their CO_2 loading, but also to a great extant by their heat capacity and viscosity. The heat capacity and viscosity opposite trend while varying MEA concentration gave rise to a maximal heating rate at a MEA concentration of around 50–60 wt%. Specific experiments were also carried out to investigate the effect of the amine concentration, the regeneration temperature and the microwave initial power on regeneration performances. Results showed that among the tested temperature, 90 °C was more suitable as better performance indicators (*EC* and EC^2) were obtained. Concerning the initial microwave power, it was found that a minimal heating power of 100 W was needed. With higher powers, one can however benefit from higher volumetric heating rates of rich solutions to slightly reduce energy consumption as CO_2-loaded solutions have lower heat capacities.

Finally, due to its high heating, absorption and regeneration rates and its good physico-chemical properties, the 50 wt% MEA concentration was found to give the best performances from cyclic experiments performed under various conditions. Under optimal conditions, a 50 wt % MEA solution offered a 13% higher CO_2 cyclic capacity and a 15% energy consumption reduction in comparison to the well-known 30 wt % concentration. These results indicate that the use of a 50 wt% MEA solution may potentially be interesting to increase cyclic capacity and reduce energy consumption in an industrial CO_2 capture process using microwave to regenerate the solutions. Of course, more works are required to evaluate other important absorbent characteristics like the degradation and corrosiveness of that solution. Furthermore, a thorough study concerning the comparison of microwave irradiation to a conventional heating method is currently in progress in our laboratories and will hopefully bring even more insight on microwave irradiation mode of action during the regeneration of spent aqueous amine solutions.

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Table 4
Results from cyclic experiments with various rich and lean CO_2 loadings.

MEA concentration (wt%)	Abs./Reg. times (min)	Rich loading	Lean loading	Cyclic capacity (mol CO_2)	Energy (J)	*EC* (kJ/mol)	EC^2 (kJ/mol^2)
30	23/16	0.49	0.27	0.0054	10706	1983	367147
	17/16	0.46	0.26	0.0047	11250	2373	500721
	11/16	0.38	0.21	0.0041	11309	2758	672754
	8/16	0.18	0.06	0.0030	11513	3838	1279222
50	23/16	0.44	0.30	0.0057	8942	1574	277165
	17/16	0.40	0.28	0.0052	9113	1753	337019
	11/16	0.36	0.26	0.0042	9500	2262	538549
	8/16	0.28	0.20	0.0031	10155	3286	1063562

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